Development of β-$(Al_xGa_{1-x})_2O_3$ /Ga_2O_3 Heterostructures

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***Abstract*— The β-$(Al_xGa_{1-x})_2O_3$/ Ga_2O_3 heterostructure is a promising element of advanced designs to fully exploit the excellent material properties of β-Ga_2O_3 for power devices. However, β-$(Al_xGa_{1-x})_2O_3$ growth is challenging because of the different crystal structure of the corundum-phase Al_2O_3 and the monoclinic phase β-Ga_2O_3. Existing growth conditions have resulted in an Al composition lower than x= 25% achieved in Molecular Beam Epitaxy (MBE) growth. A recent development of a metal-oxide catalyzed epitaxy (MOCATAXY) growth condition enabled a tremendous expansion of the growth window, pushing the growth temperature more than 200 °C higher than the typical growth temperatures. This makes it possible to achieve high Al content near 60% in the monoclinic phase based on the phase diagram. Tremendous research opportunities are foreseen in the development of high Al content β-$(Al_xGa_{1-x})_2O_3$ films. Expanding the growth regime through MOCATAXY using In could help improve heterostructure growth for future devices.**

Keywords—Ga_2O_3; heterostructure; molecular beam epitaxy; 2DEG, MOCATAXY

I. Introduction

Beta-phase gallium oxide (β-Ga_2O_3) was recently recognized as a promising candidate for electronic device applications because of its excellent intrinsic material properties, including the wide band gap energy (4.8 eV), the projected high breakdown field near 8 MV/cm and the feasibility for controllable n-type doping and contacts.[1] This leads to high figure of merit for power switching applications. Moreover, the availability of high quality single-crystal material using melt-based techniques[2] is beneficial for mass production of high quality bulk films with low defect density, which makes β-Ga_2O_3 especially attractive for power electronics applications when compared to other wide bandgap semiconductors, such as SiC and GaN. Alloying of In and Al to β-Ga_2O_3 enables further band gap tunability and provides flexibilities in device design using heterostructures.

First principle calculations suggested a staggered band alignment with a positive conduction band offset and small valence band offset between monoclinic phase $(Al_xGa_{1-x})_2O_3$ (AlGO) and β-Ga_2O_3 (GO).[3] Carrier confinement is therefore feasible at the AlGO/GO heterointerface. When a thin layer of dopants is placed in the AlGO layer, the free carriers could be transferred to the AlGO/GO interface, and thereby forming a high density two-dimensional electron gas (2DEG) through modulation doping as shown in Fig. 1. The spatial separation between the ionized impurities and the 2DEG channel can lead to significant improvement in the transport properties.[4] At the same time, the high density 2DEG could lead to enhanced screening of the scattering centers, making it possible to achieve higher electron mobilities that exceed theoretical limits in bulk doped films. Theoretical calculations[5] indicated a 2DEG mobility exceeding 500 cm^2/Vs at a charge density higher than 5×10^{12} cm^{-2}, while the bulk mobility has been demonstrated both theoretically and experimentally to be less than 180 cm^2/Vs due to strong polar optical phonon scattering[6-8]. This intrigued significant research interest on the modulation doped structures.

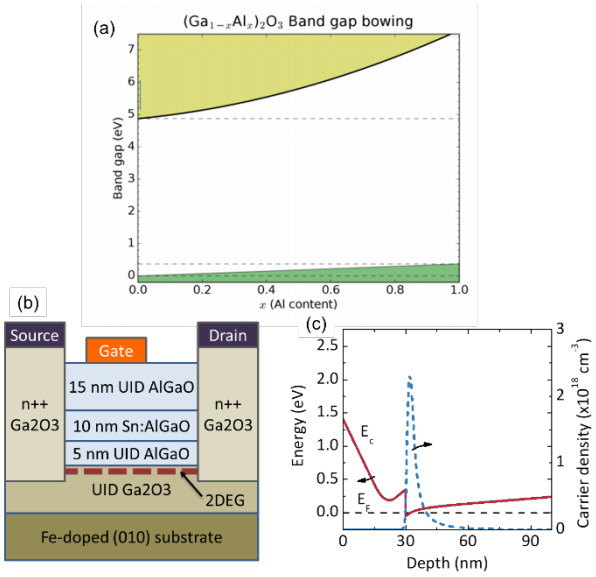

Figure 1 (a) Calculated conduction and valence band edges for β-$(AlGa)_2O_3$. Reprinted after Appl. Phys. Lett. 112, 242101 (2018)[3] (b) Schematic MODFET structure, (c) Equilibrium energy band diagram and charge profile under the gate of a MODFET.

II. Growth Challenges of AlGaO

The β-$(Al_xGa_{1-x})_2O_3$/Ga_2O_3 (AlGO/GO) heterostructures have been mainly investigated using oxygen plasma-assisted molecular beam epitaxy (PAMBE), focusing on producing high quality, coherent β-AlGO/GO heterostructures with high Al content. The initial demonstration and analysis of coherent

epitaxially grown (010) β-$(Al_xGa_{1-x})_2O_3$ was done by Kaun et al.[9] Further X-ray and compositional analysis was performed by Oshima et al[10], who derived a linear dependence of the Al composition on the separation between the (020) peaks of the AlGO and GO layers[10]. This provides an essential guidance to the evaluation of the epitaxial heterostructures.

Since Al_2O_3 is most stable in corundum phase, it is challenging to alloy between Al_2O_3 and monoclinic β-Ga_2O_3 to achieve monoclinic phase AlGO. While high quality AlGO films with sharp heterointerfaces have been demonstrated following growth optimizations, significant challenges were recognized for the realization of high Al contents while sustaining single crystal monoclinic structure in the AlGO layers. The maximum Al content has been shown to depend on growth temperature with a maximum of ~16% at 600 °C and concentrations of 25% achieved at 725 °C for fully coherent, single crystalline β-$(Al_xGa_{1-x})_2O_3$.[9] Attempts for higher Al compositions resulted in severe phase segregation in the AlGaO layer. As a result, the conduction band offset at the AlGO/GO interface is limited to be less than 0.3 eV as shown in Fig. 1(a), which further limits the maximum 2DEG density to be below 2×10^{12} cm^{-2} without forming a parasitic channel in the modulation doped structure.[4]

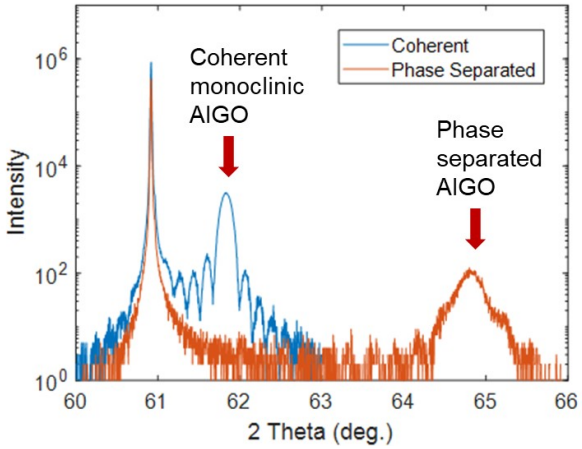

Figure 2. High resolution x-ray diffraction (HRXRD) of the AlGaO/GaO films with varying Al content in the AlGO layer.

The dependence of the maximum Al incorporation on growth temperature is consistent with the Al_2O_3-Ga_2O_3 phase diagram produced by ceramic analysis by Hill et al. in 1952 [11], shown in Fig. 3. This strong agreement with the expected thermodynamically stable phases is an interesting feature of β-$(Al_xGa_{1-x})_2O_3$ growth, but also makes it difficult to achieve high Al contents at typical (010) β-$(Al_xGa_{1-x})_2O_3$ growth temperatures of 600 – 750 °C. The phase diagram predicts a congruent point around 800 °C above which Al contents of more than 60% can be stable in the β phase. Achieving a high Al content is essential to high conduction band offset and therefore better heterostructure devices, but the conventional β-$(Al_xGa_{1-x})_2O_3$ PAMBE growth regime is limited to temperatures of 600 – 750 °C because of the reduced growth rate at high temperatures due to suboxide formation.[12]

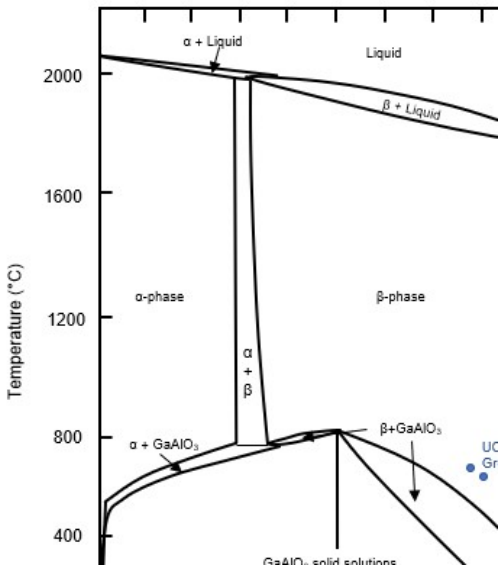

Figure 3. Al_2O_3-Ga_2O_3 Phase Diagram based on data from Hill et al.[30] along with UCSB PAMBE data.

III. MOCATAXY GROWTH

Recently, the use of In-catalyzed growth has demonstrated a remarkable expansion of the PAMBE growth regime available for β-Ga_2O_3. This involves an exchange mechanism between Ga and In in In_2O_3 formed at the growth surface with a supplied In flux[13, 14]. Ga favorably incorporates over In and at high Ga fluxes almost no In is incorporated into the single crystalline (010) β-Ga_2O_3. Through this mechanism, growth can be performed at higher temperatures and higher Ga fluxes while achieving higher growth rates that are less limited by Ga_2O suboxide desorption. This Metal Oxide Catalyzed Epitaxy (MOCATAXY) can be expanded to β-$(Al_xGa_{1-x})_2O_3$ growth as shown in Fig. 4.

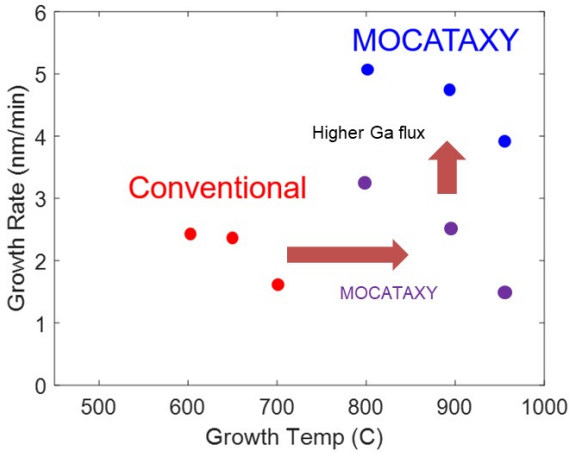

Figure 4 Dependence of Ga2O3 growth rate on the growth temperatures for conventaionl growths and MOCATAXY growths

With the Al-O bond being even stronger than Ga-O and In-O bonds, Al can favorably incorporate over the other group III metals allowing for the growth of β-$(Al_xGa_{1-x})_2O_3$ with very

limited In incorporation as demonstrated by Vogt et al.[13, 14] Despite supplying an In flux that was higher than the Al flux in III-rich conditions, β-$(Al_xGa_{1-x})_2O_3$ was still formed. APT confirmed random distribution of Al with only about 1% In incorporation despite similar Ga and In fluxes during growth. The compositions of constituents on the group III sites was found to be uniform in the films. This expands the range of growth temperatures at which coherent β-$(Al_xGa_{1-x})_2O_3$ /Ga_2O_3 to > 900 °C and even improves the growth rate. The mechanism for the improved growth rate is still not fully understood, but the formation of In_2O_3 seems to access more oxygen (likely molecular O_2 in addition to the plasma-created atomic O), which is the limitation of growth rates as III fluxes are easily increased with increasing cell temperatures. Furthermore, suboxide desorption is limited at higher growth temperatures with In. In_2O_3 could grow from molecular oxygen whereas β-Ga_2O_3 requires atomic oxygen to form, however further studies of In catalyzed growth are needed. β-Ga_2O_3 has been shown to not form without striking the plasma, suggesting that it requires atomic oxygen to grow.

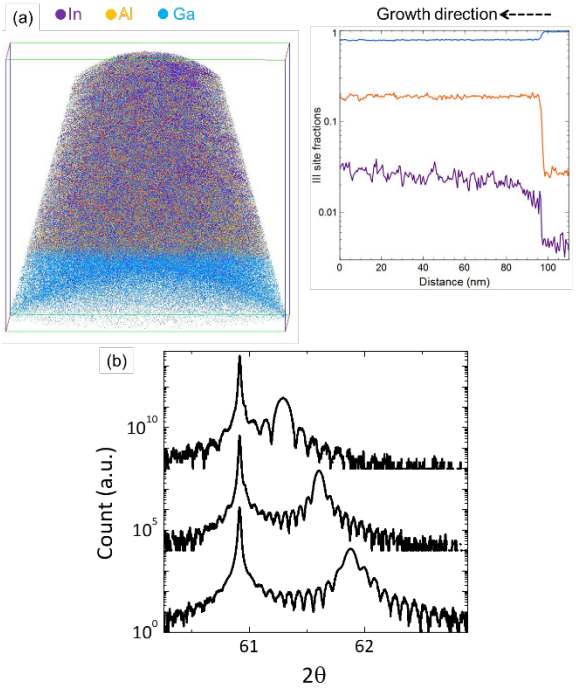

Figure 5 (a) Atom Probe Tomography of MOCATAXY AlGaO. The growth resulted in low In incorporation below 1%. (b) HRXRD of MOCATAXY (020) β-$(Al_xGa_{1-x})_2O_3$ / β-Ga_2O_3 heterostructures grown at 830 C. The Al compositions are 15%, 18% and 22%.

Coherent β-$(Al_xGa_{1-x})_2O_3$ has been demonstrated with Al contents of 25%, however much work into exploring the highly complex quaternary growth space involving In, Al, Ga, and O to improve maximum Al content still needs to be performed. The heterostructure was coherent and of high quality, demonstrated by High Resolution X-ray Diffraction (HRXRD) rocking curve full widths at half maximum (FWHMs), similar to those of the bare substrate and thickness fringes in ω-2θ scans as shown in Fig. 5. In addition, cross-sectional transmission electron microscopy (TEM) analysis revealed no evidence of dislocations and twinning. Surface morphologies were good with RMS roughness of 0.3 nm and no observed step-bunching.

IV. N-TYPE DOPING

Controllable doping of the epitaxial films is necessary for device applications. For n-type doping in PAMBE of β-Ga_2O_3, the dopant should ideally be robust to various growth conditions, easily controllable, and be easy to use in the high vacuum chamber without oxidizing in the growth environment. Sn, Si, and Ge have all been shown to be shallow donors in β-Ga_2O_3 and potential options for doping of PAMBE grown films. As a source, Si shows the most difficulty due to oxidation of the solid Si in the high background oxygen environments in PAMBE growth (i.e., the chamber pressure during growth is ~10^{-5} torr – nearly all due to molecular oxygen). However, successful pulsed delta doping has been used to realize high n-type doping concentrations and employed in various devices.[15] This scheme involves only opening the Si shutter for about a few seconds over the span of a minute and continuing this throughout the growth of a doped layer. This limits the Si source's exposure to oxygen while obtaining large fluxes of Si over a short time to achieve a target average doping concentration. Modulation doped channels with carrier density less than 2×10^{12} cm^{-2} and improved transport properties have been realized using Si delta-doping.[4]

Because of the poor doping uniformity and the ability to realize low doping concentrations using Si as dopant in oxide MBE systems, significant challenges arise for a range of device applications. Sn and Ge have demonstrated a wider range of doping achievable with Ge being the most investigated in (010) β-Ga_2O_3 PAMBE.[16] Initial attempts to dope β-$(Al_{0.2}Ga_{0.8})_2O_3$ with Sn have been able to produce doping concentrations on the order of 3 x 10^{18} cm^{-3}, which is desirable for MODFET structures and further suggests Sn's capability as a shallow donor for PAMBE growth.

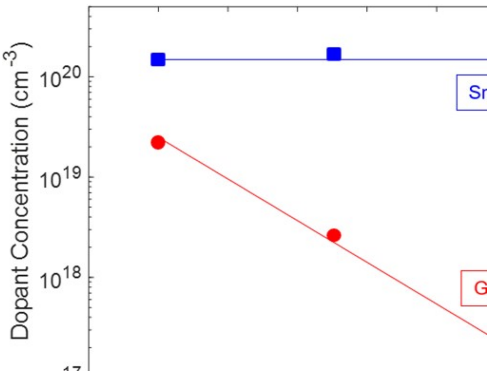

Figure 6 Dependence of doping concentration on the growth temperatures for Sn and Ge dopants.

Ahmadi et al. investigated doping with Ge over various growth regimes.[16] Ge incorporation into the films had a high dependence on growth temperature with Ge concentration dropping from 10^{19} cm^{-3} to 10^{17} cm^{-3} for a growth temperature increase from 600 °C to 700 °C. Furthermore, the Ge incorporation depends strongly on growth regime with oxygen

rich growth yielding much higher Ge doping levels of greater than 10^{20} cm^{-3} for conditions that would achieve 10^{18} cm^{-3} in metal rich growth regime. In comparison, Sn doping was also shown to achieve a wide range of doping concentrations from 5 x 10^{17} cm^{-3} to 10^{20} cm^{-3} achieving a mobility of 120 $cm^2V^{-1}s^{-1}$ for the lowest doping conditions. Sn doping showed robustness to growth temperature and group III flux in the typical growth regimes of (010) β-Ga_2O_3 as shown in Fig. 6. This makes it appealing for its application to growth at varying growth conditions. For MOCATAXY growth, effective doping concentrations above 10^{18} cm^{-3} was realized regardless of the significantly higher growth temperatures, making it feasible to achieve modulation doped structures using Sn as n-type dopant.

V. Future challenges and opportunities

Even though the studies on β-$(Al_xGa_{1-x})_2O_3$/Ga_2O_3 heterostructures are still in the early stages, tremendous progress has been made, including the optimized growth conditions for sharp interfaces and the demonstrations of modulation doping. However, several challenges remain to be explored for future studies:

1. While β-$(Al_xGa_{1-x})_2O_3$ is expected to be stable in monoclinic phase up to Al composition of ~ 60% based on the phase diagrams, successful growth of β-$(Al_xGa_{1-x})_2O_3$ films with high Al content has not been realized. The MOCATAXY growths provide a tremendous growth space to explore. At the same time, the mechanism behind the catalytic growth mode is not fully understood. Better growth control of the β-$(Al_xGa_{1-x})_2O_3$ films is expected base on further understanding of the growth kinetics. This provides great research opportunities in both materials science and device development for β-Ga_2O_3.
2. Understanding of the band structures of β-$(Al_xGa_{1-x})_2O_3$ is essential for the heterostructure device applications. However, the fundamental properties, including the conduction band offset, band gap energy, and impurity states, have not been investigated systematically. Evaluation of those properties relies on the growth of high crystalline films.
3. Theoretical calculations predicted high electron mobility in a high density 2DEG channel because of the screen of the scattering centers. Improved mobility above 500 cm^2/Vs would enable both high power and high frequency device applications. Realization of the high 2DEG charge density and experimental exploration of the transport properties could be another challenge for the heterostructures.

To summarize, the modulation doped structures could be promising for electronic device applications. The MODFETs are currently limited by the low conduction band offset due to low Al incorporation in the β-$(Al_xGa_{1-x})_2O_3$ films. There is great potential for improvement of β-Ga_2O_3 and β-$(Al_xGa_{1-x})_2O_3$ growth by MBE. The MOCATAXY growth condition provides a broad window for future investigation of β-$(Al_xGa_{1-x})_2O_3$ films with high Al content. At the same time, the possible n-type dopants will remain to be evaluated based on different device applications. The development of novel growth conditions and the exploration of n-type doping are expected to enable heterostructures that are suitable for high performance electronic device applications.

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